

Calcium Carbonate Effects on U(VI) Sorption to Soils

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Abstract: Uranium contamination of the subsurface resulting from past radioactive waste disposal and storage practices represents a serious environmental contamination problem at a number of sites. Under oxidizing geochemical conditions, the mobility of U(VI) in subsurface is primarily controlled by sorption. In many soil systems, calcite represents an important mineral phase which likely affects U(VI) sorption behavior. To quantify this influence, sorption of U(VI) to soils in the presence of naturally occurring calcium carbonate was investigated by conducting batch experiments in which either U(VI) concentration or solution pH was varied. Two soils containing different calcite concentrations were selected, one from Oak Ridge, Tennessee, and another from Altamont Pass, California. The results show that the presence of calcium carbonate in soils strongly affects U(VI) sorption. Higher concentrations of soil calcium carbonate lead to a pronounced suppression of the pH-dependent sorption curve in neutral pH range, because of the formation of a very stable neutral complex of calcium uranyl carbonate ($\text{Ca}_2\text{UO}_2(\text{CO}_3)_3$) in solution. A surface complexation model considering both strong and weak sites for ferrihydrite and ionizable hydroxyl sites for clay minerals was compared with experimental results, and U(VI) binding parameters were reasonably estimated. Fair agreement was found between the model predictions and sorption data, which span a wide range of U(VI) concentrations and pH. The results also show that selection of appropriate solution-to-solid ratios is important when measuring distribution coefficients in calcareous soils because the impact of calcium carbonate on U(VI) sorption in natural systems can be artificially diminished by dilution.